

PCT/PTO-1390 (Modified) (REV. 12-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER P 63014 US 0
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR) 09/868848
INTERNATIONAL APPLICATION NO. PCT/EP99/09850	INTERNATIONAL FILING DATE 13 DECEMBER 1999	PRIORITY DATE CLAIMED 22 DECEMBER 1998	

TITLE OF INVENTION
COLOURED GLASS SUBSTRATE CARRYING A COATING

APPLICANT(S) FOR DO/EO/US
HECQ, Andre et al

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below.
4. ☐ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
 - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☒ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☐ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☒ A copy of the International Search Report (PCT/ISA/210).

Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
21. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
22. ☐ Certificate of Mailing by Express Mail
23. ☐ Other items or information:

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.101) 09/868848		INTERNATIONAL APPLICATION NO. PCT/EP99/09850		ATTORNEY'S DOCKET NUMBER P 63014 US 0	
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24. The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) : <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1000.00 <input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$860.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00 <div style="text-align: right;">ENTER APPROPRIATE BASIC FEE AMOUNT =</div>				CALCULATIONS PTO USE ONLY <div style="border: 1px solid black; height: 100px; width: 100%;"></div>	
Surcharge of \$130.00 for furnishing the oath or declaration later than _____ months from the earliest claimed priority date (37 CFR 1.492 (e)). <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30				\$860.00 \$130.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	18 - 20 =	0	x \$18.00	\$0.00	
Independent claims	1 - 3 =	0	x \$80.00	\$0.00	
Multiple Dependent Claims (check if applicable). <input type="checkbox"/>				\$0.00	
TOTAL OF ABOVE CALCULATIONS =				\$990.00	
<input type="checkbox"/> Applicant claims small entity status. (See 37 CFR 1.27). The fees indicated above are reduced by 1/2.				\$0.00	
SUBTOTAL =				\$990.00	
Processing fee of \$130.00 for furnishing the English translation later than _____ months from the earliest claimed priority date (37 CFR 1.492 (f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30 +				\$0.00	
TOTAL NATIONAL FEE =				\$990.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input type="checkbox"/>				\$0.00	
TOTAL FEES ENCLOSED =				\$990.00	
				Amount to be: refunded	\$
				charged	\$

a. ☒ A check in the amount of **\$990.00** to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. **01-2520** A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING: Information on this form may become public. Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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24,765

REGISTRATION NUMBER

21 JUNE 2001

DATE

09/868848
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
US/DO

Applicant: HECQ et al
Application No: unknown
Filed: concurrently
Int'l Application No: PCT/EP99/09850
Int'l Filing Date: 13 DEC 1999
For: COLOURED GLASS SUBSTRATE
CARRYING A COATING

Assistant Commissioner for Patents
Box PCT
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

Prior to assigning an Application Number, and prior to any examination on the merits, please amend this application as follows:

09/868848-1001

In the Specification:

Page 1, prior to line 1, insert

-- CROSS-REFERENCE TO RELATED APPLICATIONS

This application is based on, and claims priority from PCT/EP99/09850 filed 13 December, 1999, and EP 98 124 370.2 filed 12 December 1998, the disclosures of which are hereby incorporated by reference.

BACKGROUND OF THE INVENTION --

Page 2, between lines 29 and 30, insert

-- SUMMARY OF THE INVENTION --

Page 10, before line 1, insert

-- DETAILED DESCRIPTION OF THE INVENTION --

In the claims:

Cancel claims 1-27 and replace them with new claims 28 through 45 which are presented on the attached pages which pages are numbered as 29 through 42.

REMARKS

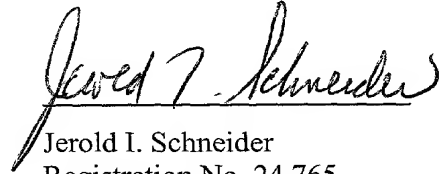
Claims 28 through 45 are pending. The amendments to the specification are to put this application into more conventional United States format.

0966044 104004

New claims are presented prior to any action by the U.S. Patent and Trademark Office.

Respectfully submitted,

June 21, 2001

A handwritten signature in cursive script, reading "Jerold I. Schneider". The signature is written in dark ink and is positioned above the printed name and address.

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U.S. PATENT & TRADEMARK OFFICE

28. Glazing comprising:

a substrate made of coloured soda-lime glass composed of main glass-forming constituents and of colouring agents, which exhibits a selectivity (LT/ET) of at least 1.1, measured with Illuminant C for a glass thickness of 4 mm; and

a pyrolytic coating deposited on the coloured glass substrate which provides the coated glazing with an increased selectivity with respect to the selectivity of the uncoated coloured glass.

29. Glazing according to Claim 28, further characterized by at least one of the following A through J, wherein:

A. the coloured glass is a glass for which the transmission between the wavelengths 1000 and 1200 nm, for a thickness of 4 mm, is lower by at least 5 points (expressed as %: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 500 and 600 nm;

B. the coloured glass is a soda-lime glass coloured dark grey composed of main glass-forming constituents and of colouring agents, in which glass the elements iron, selenium, cobalt and chromium are present as colouring agents in an amount corresponding to the following proportions (expressed as percentage by weight of the glass as if present in the form shown)

Fe_2O_3	0.75 to 1.8%
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Co	0.0040 to 0.0180%
Se	0.0003 to 0.0040%
Cr ₂ O ₃	0.0010 to 0.0100%

and the proportions of the colouring agents are such that the glass exhibits a total energy transmission, measured for a thickness of 4 mm (ET4), of between 15 and 40%, a selectivity (LTA/ET4) of at least 1.2 and an excitation purity (P) not exceeding 10%;

- C. the coating is a coating deposited by chemical vapour deposition;
- D. the coating is such that its transmission between the wavelengths 500 and 600 nm on clear glass with a thickness of 4 mm is higher by at least 3 points (expressed as percentage: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 1000 and 1200 nm;
- E. the coating is chosen from one of the following (i) through (v):
 - (i) oxide coating deposited by pyrolysis comprising tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5,
 - (ii) coating deposited by pyrolysis comprising a conductive or semiconductive layer with a thickness of 15 to 500 nm formed from a material comprising a metal oxide comprising a doping agent in a ratio of 5 to 100 mol per 100 mol of metal oxide, the metal oxide being selected from one or more of the following: tungsten

(iii) coating deposited by pyrolysis which comprises an anti-reflective interferential stacking comprising, from the glass, a stacking of materials with alternatively high and low refractive indices,

(v) titanium nitride coating deposited by pyrolysis;

- F. the coated substrate is bent and/or heat treated, in particular annealed or tempered;
- G. the light reflection factor (LR) is less than 13%;
- H. the dominant transmitted wavelength in the visible spectrum of the coated substance is less than the dominant transmitted wavelength of the uncoated substrate;
- I. the coating deposited on the coloured glass is such that if applied to 4mm thick clear glass the so coated glass would have a light transmission factor measured with Illuminant C of less than or equal to 65%; and
- J. the glazing is for a vehicle of the motor vehicle or train type.

30. Glazing according to claim 29 further characterized by at least two of the features A through J.

31. Glazing according to claim 29 further characterized by all of the features A through J.

32. Glazing according to Claim 28, further characterized by at least one of the following A through J, wherein:

A. the coloured glass is a glass for which the transmission between the wavelengths 1000 and 1200 nm, for a thickness of 4 mm, is lower by at least 5 points (expressed as %: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 500 and 600 nm;

B. the coloured glass is a green-coloured soda-lime glass which comprises the following percentages by weight of colouring agents, the total amount of iron being expressed in the form of Fe_2O_3 :

Fe_2O_3	0.7 to 1.3%
FeO	0.18 to 0.27%
Co	0 to 0.0040%
V_2O_5	0.0050 to 0.1%

and which exhibits, under Illuminant A and for a glass thickness of 4 mm, a light transmission (LTA4) of between 40 and 70%, and a selectivity (LTA/ET4) of greater than or equal to 1.50;

- C. the coating is a coating deposited by chemical vapour deposition;
- D. the coating is such that its transmission between the wavelengths 500 and 600 nm on clear glass with a thickness of 4 mm is higher by at least 3 points (expressed as percentage: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 1000 and 1200 nm;
- E. the coating is chosen from one of the following (i) through (v):
 - (i) oxide coating deposited by pyrolysis comprising tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5,
 - (ii) coating deposited by pyrolysis comprising a conductive or semiconductive layer with a thickness of 15 to 500 nm formed from a material comprising a metal oxide comprising a doping agent in a ratio of 5 to 100 mol per 100 mol of metal oxide, the metal oxide being selected from one or more of the following: tungsten oxide (WO_3) molybdenum trioxide (MoO_3), niobium pentoxide (Nb_2O_5), tantalum pentoxide (Ta_2O_5), vanadium pentoxide (V_2O_5) and vanadium dioxide (VO_2),
 - (iii) coating deposited by pyrolysis which comprises an anti-reflective interferential stacking comprising, from the glass, a stacking of materials with alternatively high and low refractive indices,

- (iv) coating which comprises a layer with an emissivity of less than 0.3, in particular a layer deposited by pyrolysis based on fluorine-doped tin oxide, and
 - (v) titanium nitride coating deposited by pyrolysis;
- F. the coated substrate is bent and/or heat treated, in particular annealed or tempered;
 - G. the light reflection factor (LR) is less than 13%;
 - H. the dominant transmitted wavelength in the visible spectrum of the coated substance is less than the dominant transmitted wavelength of the uncoated substrate;
 - I. the coating deposited on the coloured glass is such that if applied to 4mm thick clear glass the so coated glass would have a light transmission factor measured with Illuminant C of less than or equal to 65%; and
 - J. the glazing is for a vehicle of the motor vehicle or train type.

33. Glazing according to claim 32 further characterized by at least two of the features A through J.

34. Glazing according to claim 32 further characterized by all of the features A through J.

35. Glazing according to Claim 28, further characterized by at least one of the following A through J, wherein:

- A. the coloured glass is a glass for which the transmission between the wavelengths 1000 and 1200 nm, for a thickness of 4 mm, is lower by at least 5 points (expressed as %: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 500 and 600 nm;
- B. the coloured glass is a grey-green soda-lime glass composed of main glass-forming constituents and of colouring agents which comprises less than 0.4% by weight of FeO and from 0.9 to 1.8% of Fe₂O₃, which has an excitation purity of more than 5% and which exhibits, under Illuminant A and for a glass thickness of 4 mm, a light transmission (LTA4) of greater than 30%, a selectivity (LTA/ET) of greater than 1.55 and an ultraviolet radiation transmission (UVT4) of less than 10%;
- C. the coating is a coating deposited by chemical vapour deposition;
- D. the coating is such that its transmission between the wavelengths 500 and 600 nm on clear glass with a thickness of 4 mm is higher by at least 3 points (expressed as percentage: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 1000 and 1200 nm;
- E. the coating is chosen from one of the following (i) through (v):

(i) oxide coating deposited by pyrolysis comprising tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5,

(ii) coating deposited by pyrolysis comprising a conductive or semiconductive layer with a thickness of 15 to 500 nm formed from a material comprising a metal oxide comprising a doping agent in a ratio of 5 to 100 mol per 100 mol of metal oxide, the metal oxide being selected from one or more of the following: tungsten oxide (WO_3) molybdenum trioxide (MoO_3), niobium pentoxide (Nb_2O_5), tantalum pentoxide (Ta_2O_5), vanadium pentoxide (V_2O_5) and vanadium dioxide (VO_2),

(iii) coating deposited by pyrolysis which comprises an anti-reflective interferential stacking comprising, from the glass, a stacking of materials with alternatively high and low refractive indices,

(iv) coating which comprises a layer with an emissivity of less than 0.3, in particular a layer deposited by pyrolysis based on fluorine-doped tin oxide, and

(v) titanium nitride coating deposited by pyrolysis;

F. the coated substrate is bent and/or heat treated, in particular annealed or tempered;

G. the light reflection factor (LR) is less than 13%;

H. the dominant transmitted wavelength in the visible spectrum of the coated substance is less than the dominant transmitted wavelength of the uncoated substrate;

- I. the coating deposited on the coloured glass is such that if applied to 4mm thick clear glass the so coated glass would have a light transmission factor measured with Illuminant C of less than or equal to 65%; and
- J. the glazing is for a vehicle of the motor vehicle or train type.

36. Glazing according to claim 35 further characterized by at least one of the features A through J.

37. Glazing according to claim 35 and further characterized by all of the features A through J.

38. Glazing according to Claim 28, further characterized by at least one of the following A through J, wherein:

- A. the coloured glass is a glass for which the transmission between the wavelengths 1000 and 1200 nm, for a thickness of 4 mm, is lower by at least 5 points (expressed as %: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 500 and 600 nm;
- B. the coloured glass is a coloured soda-lime glass composed of main glass-forming constituents and of colouring agents which comprises from 0.40 to 0.52% by weight of FeO and which exhibits, under Illuminant A and for a glass

thickness of 4 mm, a light transmission (LTA4) of less than 70%, a selectivity (LTA/ET4) of greater than 1.65 and an ultraviolet radiation transmission (UVT4) of less than 8%;

- C. the coating is a coating deposited by chemical vapour deposition;
- D. the coating is such that its transmission between the wavelengths 500 and 600 nm on clear glass with a thickness of 4 mm is higher by at least 3 points (expressed as percentage: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 1000 and 1200 nm;
- E. the coating is chosen from one of the following (i) through (v):
 - (i) oxide coating deposited by pyrolysis comprising tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5,
 - (ii) coating deposited by pyrolysis comprising a conductive or semiconductive layer with a thickness of 15 to 500 nm formed from a material comprising a metal oxide comprising a doping agent in a ratio of 5 to 100 mol per 100 mol of metal oxide, the metal oxide being selected from one or more of the following: tungsten oxide (WO_3) molybdenum trioxide (MoO_3), niobium pentoxide (Nb_2O_5), tantalum pentoxide (Ta_2O_5), vanadium pentoxide (V_2O_5) and vanadium dioxide (VO_2),

(iii) coating deposited by pyrolysis which comprises an anti-reflective interferential stacking comprising, from the glass, a stacking of materials with alternatively high and low refractive indices,

(iv) coating which comprises a layer with an emissivity of less than 0.3, in particular a layer deposited by pyrolysis based on fluorine-doped tin oxide, and

(v) titanium nitride coating deposited by pyrolysis;

F. the coated substrate is bent and/or heat treated, in particular annealed or tempered;

G. the light reflection factor (LR) is less than 13%;

H. the dominant transmitted wavelength in the visible spectrum of the coated substance is less than the dominant transmitted wavelength of the uncoated substrate;

I. the coating deposited on the coloured glass is such that if applied to 4mm thick clear glass the so coated glass would have a light transmission factor measured with Illuminant C of less than or equal to 65%; and

J. the glazing is for a vehicle of the motor vehicle or train type.

39. Glazing according to claim 38 further characterized by at least two of the features A through J.

40. Glazing according to claim 38 further characterized by all of the features A through J.
41. Glazing according to claim 28 further characterized by one of the following:
- A. the selectivity of the uncoated coloured glass is at least 1.3;
 - B. the selectivity of the uncoated coloured glass is less than or equal to 2.
42. Glazing according to claim 28 characterized in that the selectivity of the coated substrate is greater than 2.
43. Glazing according to Claim 28, further characterized by one of the following:
- A. the selectivity is increased by at least 3% with respect to the selectivity of the uncoated coloured glass;
 - B. the selectivity is increased by at least 10% with respect to the selectivity of the uncoated coloured glass.

44. Glazing according to claim 28, characterized by one of the following:
- A. the coating comprises an underlayer between the substrate and the coating deposited by pyrolysis;
 - B. that the coating is an oxide coating which is preferably deposited by vapour-phase pyrolysis comprises tin and antimony in a molar ratio Sb/Sn of between 0.04 and 0.16;
 - C. the coating is an oxide coating deposited by pyrolysis which comprises tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5 and its thickness is between 250 and 500 nm.
45. Glazing according to claim 28, characterized by one of the following:
- A. the coating deposited on the coloured glass is such that if applied to 4 mm thick clear glass the so coated glass would have a light transmission factor measured with illuminant C of less than or equal to 70%;
 - B. the coating deposited on the coloured glass is such that if applied to 6 mm thick clear glass the so coated glass would have a light transmission factor measured with illuminant C of less than or equal to 65%;

- C. the light transmission factor of the coated glazing is less than the light transmission factor of the uncoated glass substrate by a factor of greater than 10% calculated according to the formula

$$\frac{LT_{\text{uncoated substrate}} - LT_{\text{coated glazing}}}{LT_{\text{uncoated substrate}}} \times 100.$$

- D. the light transmission factor of the coated glazing is less than the light transmission factor of the uncoated glass substrate by a factor of greater than 15% calculated according to the formula

$$\frac{LT_{\text{uncoated substrate}} - LT_{\text{coated glazing}}}{LT_{\text{uncoated substrate}}} \times 100.$$

Coloured glass substrate carrying a coating

The present invention relates to a glazing comprising:

- a substrate made of coloured soda-lime glass composed of main glass-forming constituents and of colouring agents,
- a coating deposited on the coloured glass substrate.

The expression "soda-lime glass" is used here in the broad sense and relates to any glass which comprises the following constituents (percentages by weight):

	SiO ₂	60 to 75%
	Na ₂ O	10 to 20%
10	CaO	0 to 16%
	K ₂ O	0 to 10%
	MgO	0 to 10%
	Al ₂ O ₃	0 to 5%
	BaO	0 to 2%
15	BaO + CaO + MgO	10 to 20%
	K ₂ O + Na ₂ O	10 to 20%.

This type of glass finds very wide use in the field of glazings for motor vehicles or buildings, for example. It is commonly manufactured in the form of a ribbon by the float-glass process. Such a ribbon can be cut in the form of sheets, which can subsequently be bent or subjected to a treatment for enhancing the mechanical properties, for example a thermal tempering treatment.

When referring to the optical properties of a glass sheet, it is generally necessary to relate these properties to a standard illuminant. In the present description, two standard illuminants are used: Illuminant C and Illuminant A defined by the Commission Internationale de l'Eclairage (C.I.E.) [International Commission on Illumination]. Illuminant C represents average daylight having a colour temperature of 6700 K. This illuminant is especially of use in evaluating the optical properties of glazings intended for buildings. Illuminant A represents the radiation from a Planck radiator at a temperature of approximately 2856 K. This illuminant represents the light emitted by car headlamps and is especially intended to evaluate the optical properties of glazings intended for motor vehicles.

The "light transmission factor" (LT) is the percentage of incident luminous flux transmitted in the visible region, between 380 and 780 nm, through a substrate.

The "light reflection factor" (LR) is the percentage of incident luminous flux reflected by a substrate.

The "ultraviolet transmission factor" (UVT4) is the total percentage of transmission in the ultraviolet, between 290 nm and 380 nm, for a glass with a thickness of 4 mm.

The "energy transmission factor" (ET) is the total percentage of incident energy radiation directly transmitted through a substrate between the wavelengths 300 and 2500 nm.

The "solar factor" (SF) is the sum of the total energy directly transmitted through a substrate (ET) and of the energy which is absorbed (AE) and reradiated at the face opposite the energy source, with respect to the total energy radiation reaching the substrate.

The "selectivity" of a coated substrate represents the ratio between the light transmission factor and the energy transmission factor. It can be defined as being the ratio of the light transmission factor to the solar factor (LT/SF), which is used in particular in the case of a building glazing. It can also be defined as being the ratio of the light transmission factor to the energy transmission factor (LT/ET), in particular in the case of glazings for vehicles, for which the energy which is absorbed and reradiated is regarded as negligible when the vehicle is in motion.

The "dominant wavelength" (λ_D) is the peak wavelength in the range transmitted or reflected by the coated substrate.

The "purity" (P) of the colour of the substrate refers to the excitation purity measured using Illuminant C. It is defined according to a linear scale in which a defined source of white light has a purity of 0 and the pure colour has a purity of 100%. The purity of a coated substrate is measured on the side opposite the coated face.

The "emissivity" (ϵ) is the ratio of the energy emitted by a given surface at a given temperature to that of a perfect radiator (black body having an emissivity of 1.0) at the same temperature.

From a technical viewpoint, it is desirable for the glazing, when subjected to sunshine, not to allow too great a proportion of the total incident solar radiation to pass through, in order for the interior of the vehicle or building not to become overheated.

One of the objectives of the present invention is to obtain a glazing which exhibits a high level of solar protection combined with a high selectivity and which can be obtained under conventional industrial manufacturing conditions.

It has been discovered that this objective, and others, can be achieved by a glazing which combines a selective coloured glass substrate, which

exhibits a selectivity (LT/ET) of at least 1.1, measured with Illuminant C for a glass thickness of 4 mm, and a pyrolytic coating which provides the coated glazing with an increased selectivity with respect to the selectivity of the uncoated coloured glass. In the context of the present invention, the glazing comprises a coloured glass substrate.

The present invention makes it possible to obtain a glazing exhibiting a high selectivity while avoiding the difficulties which are often associated with the industrial manufacture of coloured glasses of high selectivity. In addition, the present invention makes possible a greater flexibility in manufacturing in the sense that it is generally easier to modify the properties of the glazing by changing the coating rather than by modifying a coloured glass composition.

The coloured glass is preferably chosen so that its selectivity has a value of at least 1.3. This contributes to easily obtaining a coated glazing of high selectivity.

However, the selectivity of the uncoated coloured glass is preferably less than or equal to 2. It is possible for uncoated coloured glasses to have a selectivity of greater than 2; however, these are generally difficult to obtain under conventional industrial manufacturing conditions for two reasons: a high content of iron needed in order to obtain the high selectivity renders the glass difficult to melt, which then requires the use of particular electric furnaces, which are of low-capacity and, furthermore, a high redox ratio of FeO/total Fe which is favourable to the achievement of a high selectivity requires modifying the refining conditions, which renders the preparation of the glass problematic under industrial manufacturing conditions. This is why the selectivity of the uncoated coloured glass is preferably less than or equal to 2.

The coloured glass is preferably a glass for which the transmission between the wavelengths 1000 and 1200 nm, for a thickness of 4 mm, is lower by at least 5 points (expressed as %: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 500 and 600 nm. A coloured glass which meets this condition is selective and can in particular be green, blue or grey.

The coloured glass can be a soda-lime glass coloured dark grey composed of main glass-forming constituents and of colouring agents, in which glass the elements iron, selenium, cobalt and chromium are present as colouring agents in an amount corresponding to the following proportions (expressed as percentage by weight of the glass as if they are present in the form shown)

Fe ₂ O ₃	0.75 to 1.8%
Co	0.0040 to 0.0180%

Se	0.0003 to 0.0040%
Cr ₂ O ₃	0.0010 to 0.0100%

and the proportions of the colouring agents are such that the glass exhibits a total energy transmission, measured for a thickness of 4 mm (ET4), of
 5 between 15 and 40%, a selectivity (LTA/ET4) of at least 1.2 and an excitation purity (P) not exceeding 10%.

As an alternative form, the coloured glass is a green-coloured soda-lime glass which comprises the following percentages by weight of colouring agents, the total amount of iron being expressed in the form of Fe₂O₃:

10	Fe ₂ O ₃	0.7 to 1.3%
	FeO	0.18 to 0.27%
	Co	0 to 0.0040%
	V ₂ O ₅	0.0050 to 0.1%, and which exhibits,

under Illuminant A and for a glass thickness of 4 mm, a light transmission (LTA4) of
 15 between 40 and 70% and a selectivity (LTA/ET4) of greater than or equal to 1.50.

According to another preferred embodiment of the invention, the coloured glass is a grey-green soda-lime glass composed of main glass-forming constituents and of colouring agents which comprises less than 0.4% by weight of FeO and from 0.9 to 1.8% of Fe₂O₃, which has an excitation purity of more than
 20 5% and which exhibits, under Illuminant A and for a glass thickness of 4 mm, a light transmission (LTA4) of greater than 30%, a selectivity (LTA/ET) of greater than 1.55 and an ultraviolet radiation transmission (UVT4) of less than 10%.

As an alternative form, the coloured glass is a coloured soda-lime glass composed of main glass-forming constituents and of colouring agents which
 25 comprises from 0.40 to 0.52% by weight of FeO and which exhibits, under Illuminant A and for a glass thickness of 4 mm, a light transmission (LTA4) of less than 70%, a selectivity (LTA/ET4) of greater than 1.65 and an ultraviolet radiation transmission (UVT4) of less than 8%.

A particularly pleasing aesthetic appearance in combination with a
 30 high selectivity can also be obtained with a coloured soda-lime glass with a blue hue composed of main glass-forming constituents, including more than 2% of magnesium oxide, and of colouring agents which comprises more than 1.1% by weight of Fe₂O₃, less than 0.53% by weight of FeO and less than 0.13% of manganese oxide, which has a light transmission (LTA4) of between 15% and 70%
 35 and a selectivity (SE4) of greater than 1.2, and which exhibits a dominant wavelength (λ_D) and a purity (P) such that they lie in a CIE trichromatic diagram inside a triangle, the tips of which are defined by the points representing Illuminant C and the points with the (λ_D , P) coordinates (490,19) and (476,49) respectively.

In order to obtain a higher value of selectivity and/or a significant improvement in the selectivity with respect to the starting uncoated coloured glass, the selectivity is preferably increased by at least 3% with respect to the selectivity of the uncoated coloured glass. The selectivity is preferably increased by at least 10% with respect to the selectivity of the uncoated coloured glass.

Various techniques are known for forming coatings on a substrate made of vitreous material, including pyrolysis and cathodic sputtering. The coating of the present invention is a coating deposited by pyrolysis. Pyrolysis generally exhibits the advantage of forming a hard coating which does not require a protective layer. Coatings formed by pyrolysis exhibit lasting properties of resistance to abrasion and corrosion. It is believed that this is due in particular to the fact that the process comprises the deposition of precursor on a substrate which is hot. Furthermore, pyrolysis is generally less expensive than other coating processes, such as cathodic sputtering, particularly in terms of investment in equipment. This is because deposition by pyrolysis can be carried out continuously during the manufacture of the glass on the freshly formed hot glass ribbon. In contrast, a deposition such as by cathodic sputtering is carried out under vacuum away from the manufacturing line on cold glass cut up beforehand into sheets. In addition, the deposition of coating by processes other than pyrolysis, for example by cathodic sputtering, results in products which exhibit different properties, in particular a decreased resistance to abrasion and sometimes different optical properties (refractive index).

The coating deposited by pyrolysis can be applied to the substrate by chemical vapour deposition or starting from a liquid precursor. The coating (or the coating layers) is preferably applied to the substrate by chemical vapour deposition. Chemical vapour deposition is particularly preferred because it tends to produce coatings with an even thickness and composition, the uniformity of the product being particularly important when the glazings have to be used for large surface areas. With the use of liquid reactive materials, it is not possible to influence the evaporation process, which renders it random. In addition, chemical vapour deposition is more economic from the viewpoint of the utilization of the starting materials, which results in decreased wastage.

The coating is preferably such that its transmission between the wavelengths 500 and 600 nm on clear glass with a thickness of 4 mm is higher by at least 3 points (expressed as percentage: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 1000 and 1200 nm.

Such a coating also makes it possible to obtain a selective glazing with clear glass.

Preferably, the coating is chosen from the following:

- 5 - coating deposited by pyrolysis comprising tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5,
- oxide coating deposited by pyrolysis comprising a conductive or semiconductive layer with a thickness of 15 to 500 nm formed from a material comprising a metal oxide comprising a doping agent in a ratio of 5 to 100 mol per 100 mol of metal oxide, the metal oxide being selected from one or more of the
10 following: tungsten oxide (WO_3), molybdenum trioxide (MoO_3), niobium pentoxide (Nb_2O_5), tantalum pentoxide (Ta_2O_5), vanadium pentoxide (V_2O_5) and vanadium dioxide (VO_2),
- coating deposited by pyrolysis which comprises an anti-reflective interferential stacking which comprises, from the glass, a stacking of materials with
15 alternatively high and low refractive indices,
- coating deposited by pyrolysis which comprises a layer with an emissivity of less than 0.3, in particular a layer deposited by pyrolysis based on fluorine-doped tin oxide
- titanium nitride coating deposited by pyrolysis. It can be in the form
20 of a stacking based on titanium nitride layer, which may be oxidised.

It has been discovered that such coatings are particularly suitable for increasing the selectivity of the glazing with respect to the selectivity of the uncoated coloured glass.

Oxide coatings deposited by pyrolysis comprising tin and antimony
25 in a molar ratio Sb/Sn of between 0.01 and 0.5 are disclosed in particular in Patent Applications GB 2,302,102 and GB 2,302,101, the contents of which are incorporated in the present application by way of reference. Such coatings provide an increase in selectivity ranging up to 20%.

Coatings deposited by pyrolysis can comprise a conductive or
30 semiconductive layer with a thickness of 15 to 500 nm formed from a material comprising a metal oxide comprising a doping agent in a ratio of 5 to 100 mol per 100 mol of metal oxide, the metal oxide being selected from one or more of the following: tungsten oxide (WO_3), molybdenum trioxide (MoO_3), niobium pentoxide (Nb_2O_5), tantalum pentoxide (Ta_2O_5), vanadium pentoxide (V_2O_5) and vanadium
35 dioxide (VO_2). Such coatings provide an increase in selectivity ranging up to 30%. In addition, they make possible a considerable increase in the selectivity in terms of LT/SF ratio: the selectivity of the glazing can be more than 30% greater than the selectivity of the uncoated coloured glass.

The coating can comprise a layer with an emissivity of less than 0.3. A layer of oxide comprising tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5 described above can possess this property. As an alternative form, the layer of low emissivity can be a layer based on fluorine-doped tin oxide. A layer with a low emissivity makes it possible to increase the selectivity in terms of LT/SF ratio, as the emissivity affects the value of the solar factor: a low emissivity makes it possible to decrease the solar factor of the glazing and thus to increase the selectivity calculated in terms of LT/SF ratio.

The coating can comprise an underlayer between the substrate and the coating deposited by pyrolysis. This underlayer can also be deposited by pyrolysis. The role of such an underlayer can be to reduce the haze by preventing the migration of sodium ions from the glass, by diffusion or by any other way, to the coating layer, whether during the formation of this upper layer or during a subsequent high-temperature treatment. For example, such an underlayer can be formed of a silicon oxide having a geometric thickness of approximately 100 nm. Such an underlayer is not generally necessary for glazings with a low light transmission factor as in such cases the haze is not apparent to a significant extent. Another role of an underlayer, as an alternative form or in addition to the role set out above, can be to form an "anti-reflecting" stacking, for example by means of an aluminium/vanadium oxidized underlayer, as disclosed in Patent Application GB 2,248,243.

In preferred embodiments of the invention, the coating is a coating deposited by pyrolysis, preferably deposited in the vapour phase, which comprises tin and antimony in a molar ratio Sb/Sn of between 0.04 and 0.16. It has been discovered that coatings comprising such proportions of tin and antimony make it possible to produce glazings, the high selectivity properties of which are particularly advantageous: the increase in selectivity of the coated glazing with respect to the uncoated coloured glass is generally at least 7% and often more than 10%.

The oxide coating deposited by pyrolysis, which comprises tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5, preferably has a thickness of between 250 and 500 nm. Such thicknesses can be obtained under conventional industrial manufacturing conditions, whereas thin or very thick coatings are difficult to produce in practice, and these thicknesses make it possible to obtain increases in selectivity which can be very large, for example more than 20%, with respect to the coloured glass alone without coating.

For some applications, it is necessary for the coated substrate to be subjected to a subsequent treatment during which it is bent and/or heat treated, in particular annealed or tempered. This is particularly the case with motor vehicle

glazings. The glazing according to the invention preferably withstands this treatment, in particular if the coating is deposited by pyrolysis. In this case, its properties are not significantly modified by the subsequent treatment. As an alternative form, if it is a coating deposited by cathodic sputtering, it is sufficient for this coating to comprise one or more so-called "sacrificial" layers which make it possible to protect the active layer or layers during the subsequent treatment.

The light reflection factor (LR) of the glazing is generally low, which is particularly advantageous for vehicle glazings. The light reflection factor (LR) of the glazing is preferably less than 13%.

The emissivity of the glazing is preferably less than 0.2. The emissivity affects the value of the solar factor: a low emissivity makes it possible to decrease the solar factor of the glazing and thus to increase the selectivity calculated in terms of LT/SF ratio. On the other hand, a low emissivity coating is generally not as good as antisolar coatings to decrease the energy transmission factor thus to increase the selectivity calculated in terms of LT/ET.

The selectivity of the coated substrate can be very high and it is preferably greater than 2. A glazing with such high selectivity is particularly desired for reasons of comfort in the field of automobile glazings.

It has been discovered that the glazing according to the invention can exhibit a dominant transmitted wavelength in the visible spectrum of the coated substrate which is less than the dominant transmitted wavelength of the uncoated substrate, preferably by at least 1, 2, 3, 4, 5 nm or even more. This is particularly advantageous when the coloured glass has a transmitted colour which is not greatly appreciated aesthetically: a typical example is green glass, having a dominant transmitted wavelength which is between 490 and 550 nm and more particularly between 490 and 520 nm. Such a glass is obtained by addition of iron to the composition of the glass. The iron makes it possible to obtain very high selectivities but the increase in the amount of iron in the composition, in combination with an increase in the selectivity, also increases the green appearance in transmission, which is generally not greatly appreciated for aesthetic reasons. The present invention makes it possible to overcome this disadvantage by providing a glazing for which the dominant transmitted wavelength is less than the dominant transmitted wavelength of the coating-free coloured glass and therefore a glass which will have a less green and more bluish appearance than the starting coloured glass, which is regarded as more desirable from an aesthetic viewpoint. It has been discovered that this advantage is particularly achieved when the coating is a coating deposited by pyrolysis comprising tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5 and more particularly a coating deposited by pyrolysis,

preferably deposited in the vapour phase, which comprises tin and antimony in a molar ratio Sb/Sn of between 0.04 and 0.16, preferably between 0.08 and 0.14. Such a glazing additionally exhibits the advantage of having advantageous energy properties because such a coating reflects in particular wavelengths situated in the far infrared, whereas the coloured glass absorbs in particular in the near infrared: there is therefore a complementary effect which is highly favourable to the selectivity of the assembly.

Consequently and preferably, the dominant transmitted wavelength in the visible spectrum of the coated substrate is less than 500 nm, preferably less than or equal to 495 nm, preferably in combination with a purity of less than 20%.

The glazing according to the invention is particularly suitable as vehicle glazing. In particular, it can be used as motor vehicle or train glazing.

One particular application of the invention is in the possibility it provides in providing an advantageous combination of properties in a glazing for solar control applications. In this respect:

- The coating may be such that if applied to 4 mm thick clear glass the so coated glass would have a light transmission factor measured with illuminant C of less than or equal to 70%, 65%, 60%, 55%, 50%, 45%, 40% or 35%.
- The coating may be such that if applied to 6 mm thick clear glass the so coated glass would have a light transmission factor measured with illuminant C of less than or equal to 65%, 60%, 55%, 50%, 45%, 40% or 35%.
- The coating may reduce the light transmission factor of the coated glazing such that the ratio of the light transmission factor of the coated glazing divided by the uncoated glass substrate is less than 0.9, 0.85, 0.8, 0.75, 0.7, 0.65, 0.6, 0.55 or 0.5 in which the light transmission factor is preferably measured using illuminant C.
- The coating may reduce the light transmission factor of the coated glazing by greater than 10%, 12%, 14%, 15%, 16%, 17%, 18%, 19%, 20%, 21%, 22%, 23%, 24%, 25%, 26%, 27%, 28%, 29%, 30%, 31%, 32%, 33%, 34%, 35%, 36%, 37%, 38%, 39% or 40% with respect to the uncoated glass substrate calculated according to the formula

$$\frac{LT_{\text{uncoated substrate}} - LT_{\text{coated glazing}}}{LT_{\text{uncoated substrate}}} \times 100.$$

in which LT is preferably measured using illuminant C.

Such advantages may be achieved using a glass substrate having a thickness of, for example about 2mm, 4mm, 6mm, 8mm, 10mm, 12mm or more.

The invention will now be described in more detail with reference to the following nonlimiting examples.

5 The properties of the various types of coloured glass are presented in Table 1 below. The properties are measured on glass samples with a thickness of 4 mm. For some glasses, the properties for a thickness of 6 mm are mentioned, in addition. The initials in this table and in the other following tables (LTA, UVT4, LTC, ET, and the like) have the meanings described above. In addition, the main colouring agents present in the various coloured glasses are mentioned in Table 1.

Table 1

Type of glass Thickness	Green 4 mm 6 mm	Dark green 4 mm 6 mm	Very dark green 4 mm 6 mm	Grey 1 4 mm	Grey 2 4 mm	Blue 1 4 mm	Blue 2 4 mm
Wavelength (III C) (nm) trans.	504 504	498 499	496 496	494	495	489	486
Purity (III C)(%)	3.5 5.2	7.7 11	11.2 15.8	5.8	5	16.8	24.3
LTA (%)	65.3 55.3	45.3 32.2	32.2 19.4	35.4	48.1	42.7	36.94
UVT4		<10 %	2.9				
LTC (%)	67 57.42	47.5 34.5	34.3 21.3	36.5	49.4	45.3	39.9
ET CIE (%)	38.6 28.4	25.2 15.7	17.7 9.5	26.3	34.6	26	21.4
SF	52.8 45.2	42.9 35.8	37.3 31.3	43.7	49.9	43.5	40.1
LTC/ET	1.74 2.02	1.88 2.20	1.94 2.24	1.39	1.43	1.74	1.86
LTC/SF	1.27 1.27	1.11 0.96	0.92 0.68	0.84	0.99	1.04	1
Colorants:							
Fe ₂ O ₃ (%)	0.94	1.32	1.55	1.4	1.13	1.26	1.24
(FeO calc %)	0.24	<0.4	0.44				
Co (ppm)	7	43	80	94	61	67	81
V ₂ O ₅ (ppm)	153	238	429	imp.	imp.	7	imp.
Cr ₂ O ₃ (ppm)	imp.	133	214	17	23	77	imp.
Se (ppm)	imp.	imp.	imp.	8	imp.	imp.	imp.

imp. = impurities

EXAMPLES 1 to 5, Comparative Examples 1 and 2

“Green”-coloured float glass as defined in Table 1, progressing along a float tank, is coated by CVD (Chemical Vapour Deposition) pyrolysis by means of a reactant comprising a mixture of MBTC (monobutyltrichlorotin) as source of tin and of SbCl_3 as source of antimony. The reactant mixture is vaporized in a stream of anhydrous air at approximately 200°C . The vaporization is facilitated by the nebulization of these reactants in the carrier gas. Superheated steam at approximately 200°C is subsequently introduced.

The coating process is continued until the geometric thickness of the tin/antimony oxide coating overlying the substrate is approximately 280 nm (Examples 1 to 3) or 440 nm (Examples 4 and 5). The Sb/Sn molar ratio in the coating, measured by X-ray fluorescence, is mentioned in the table, as well as the thicknesses, also measured by X-ray fluorescence.

As Comparative Example 1, similar glass is coated with an absorbent layer with a thickness of approximately 45 nm, deposited by pyrolysis, formed of a mixture of Fe, Co and Cr oxides in the proportions by weight 26% Fe_2O_3 , 61% Co_3O_4 and 13% Cr_2O_3 .

As Comparative Example 2, similar glass is coated with a layer of approximately 55 nm, deposited by pyrolysis, comprising titanium and tin oxides (90% TiO_2 by weight and 10% SnO_2 by weight).

Table 2 shows that, for the comparative examples, the selectivity is decreased with respect to the selectivity of the starting glass, whereas the dominant transmitted wavelength and the purity are increased, which provides an even more marked green appearance regarded as unfavourable from an aesthetic viewpoint. In contrast, for Examples 1 to 5, the selectivity is markedly increased with respect to the selectivity of the coloured glass alone: an increase of up to 19%. Furthermore, the dominant transmitted wavelength is less than that of the coloured glass alone, which is favourable from the viewpoint of the desired properties.

The light reflection (LR) and solar factor (SF) values are measured on the coated/uncoated side.

The coating of example 1 in Table 2 reduces the light transmission factor of the coated glazing by 25.4% with respect to the uncoated glass substrate calculated according to the formula:

$$\frac{LT_{\text{uncoated substrate}} - LT_{\text{coated glazing}}}{LT_{\text{uncoated substrate}}} \times 100.$$

This may be calculated as follows:

TLC of uncoated 4mm Green glass: 67% (from Table 1)

TLC of coated 4mm Green glass according to example 1:

50% (from Table 2)

Absolute reduction in TLC due to coating: $67\% - 50\% = 17$

Reduction in TLC due to coating with respect to the uncoated substrate:

5 $(17/67) \times 100 = 25.4\%$

As an alternative form, the results are given here for an identical coloured glass with a thickness of 6 mm instead of 4 mm, in Table 3.

Table 2

Example	Green glass, 4 mm, LT/ET glass alone = 1.74							
	LTC	λ_p tr	P	ET	SF	LR	LT/ET	% difference
Comparative 1	28.4	567	13	18.8	33/36	34/17	1.51	-0.23
Comparative 2	50	550	5.8	29	41/44	31/19	1.72	-0.02
1, 5% Sb/Sn 280 nm	50	502	4.3	24	41/37	9/7.5	2.08	0.34
2, 9% Sb/Sn 285 nm	42	495	6.2	21	38/35	8/7	2.00	0.26
3, 13% Sb/Sn 270 nm	37	492	7.7	19	37/34	7/6	1.95	0.21
4, 12% Sb/Sn 440 nm	27	491	10	13	33/28	6/6	2.08	0.34
5, 9% Sb/Sn 445 nm	32	491	9	15.5	34/29	7/6	2.06	0.32

Table 3

Example	Green glass, 6 mm, LT/ET glass alone = 2.02							
	LTC	λ_p tr	P	ET	SF	LR	LT/ET	% difference
Comparative 1	24	561	13	13	28/33	34/14	1.85	-0.17
Comparative 2	42.6	540	6.5	20.7	35/39	31/15	2.06	0.04
1', 5% Sb/Sn 280 nm	43	504	5.8	18.4	36/32	8.5/7	2.34	0.32
2', 9% Sb/Sn 285 nm	36	498	7.4	15.7	34/31	7.7/6	2.29	0.27
3', 13% Sb/Sn 270 nm	32	495	8.8	14	33/30	7/6	2.29	0.27
4', 12% Sb/Sn 440 nm	23	463	11.5	10	31/26	6/5	2.30	0.28
5', 9% Sb/Sn 445 nm	27	493	10	12	32/27	7/6	2.25	0.23

EXAMPLES 6 to 10

In Examples 6 to 10, the process of Examples 1 to 5 is followed but with dark green glass as defined in Table 1, with a thickness of 4 mm or, as an alternative form, of 6 mm:

Table 4:

Example	Dark green glass, 4 mm, LT/ET glass alone = 1.88							
	LTC	λ_p tr	P	ET	SF	LR	LT/ET	% difference
6, 5% Sb/Sn 280 nm	35	496	9	16.5	35/31	8/6	2.12	13%
7, 9% Sb/Sn 285 nm	30	493	11	14	33/30	7/6	2.14	14%
8, 13% Sb/Sn 270 nm	26	491	12	13	32/30	7/5	2.00	6%
9, 12% Sb/Sn 440 nm	19	491	15	9	30/25	6/5	2.11	12%
10, 9% Sb/Sn 445 nm	23	491	13	10.5	30/26	7/5	2.19	17%

Table 5:

Example	Dark green glass, 6 mm, LT/ET glass alone = 2.20							
	LTC	λ_p tr	P	ET	SF	LR	LT/ET	% difference
6', 5% Sb/Sn 280 nm	26	497	12	10.5	30/26	8/5	2.48	13%
7', 9% Sb/Sn 285 nm	22	495	14	9	29/26	7/5	2.44	11%
8', 13% Sb/Sn 270 nm	19	493	15	8	29/26	7/5	2.38	8%
9', 12% Sb/Sn 440 nm	14	492	18	6	27/23	6/5	2.33	6%
10', 9% Sb/Sn 445 nm	16	492	16	6.8	28/23	7/5	2.35	7%

It is observed that the selectivity is markedly increased: from 6 to 17% for glass with a thickness of 4 mm, and, furthermore, the dominant transmitted wavelength is less than that of the coating-free coloured glass.

EXAMPLES 11 to 15

- 5 In Examples 11 to 15, the process of Examples 1 to 5 is followed but with very dark green glass as defined in Table 1, with a thickness of 4 mm or, as an alternative form, of 6 mm:

Table 6:

Example	Very dark green glass, 4 mm, LT/ET glass alone = 1.94									
	LTC	λ_p tr	P	ET	SF	LR	LT/ET	difference	% difference	
11, 5% Sb/Sn 280 nm	25	495	13	11.5	31/27	8/5	2.14	0.20	10%	
12, 9% Sb/Sn 285 nm	21	493	14	10	30/27	7/5	2.08	0.14	7%	
13, 13% Sb/Sn 270 nm	19	491	16	9	30/27	7/5	2.00	0.06	3%	
14, 12% Sb/Sn 440 nm	14	491	18	6.4	28/23	6/5	2.00	0.06	3%	
15, 9% Sb/Sn 445 nm	16	491	17	7.4	28/23	7/5	2.07	0.13	6%	

Table 7:

Example	Very dark green glass, 6 mm, LT/ET glass alone = 2.24									
	LTC	λ_p tr	P	ET	SF	LR	LT/ET	difference	% difference	
11', 5% Sb/Sn 280 nm	16	495	17	6.3	27/22	8/5	2.54	0.30	13%	
12', 9% Sb/Sn 285 nm	13	494	19	5.4	27/23	7/5	2.41	0.17	7%	
13', 13% Sb/Sn 270 nm	12	493	20	4.8	27/23	7/5	2.50	0.26	12%	
14', 12% Sb/Sn 440 nm	8.7	492	22	3.5	26/21	6/4	2.49	0.25	11%	
15', 9% Sb/Sn 445 nm	10	492	21	4.1	26/21	7/5	2.44	0.20	9%	

The selectivity is improved and the dominant transmitted wavelength is less than that of the uncoated coloured glass. It may be noted that the percentage increase in the selectivity is less than the percentage increase obtained in the preceding examples. However, it is necessary to take into account the starting light transmission when the selectivities are compared: the lower the starting light transmission, the more difficult it is to increase the selectivity.

EXAMPLES 16 to 20

In Examples 16 to 20, the process of Examples 1 to 5 is followed but with "blue 1" glass as defined in Table 1, with a thickness of 4 mm:

Table 8:

Example	Blue 1 glass, 4 mm, LT/ET glass alone = 1.74									
	LTC	λ_p tr	P	ET	SF	LR	LT/ET	difference	% difference	
16, 5% Sb/Sn 280 nm	34	488	15	17	35/31	8/6	2.00	0.26	15%	
17, 9% Sb/Sn 285 nm	28	487	17	15	34/30	8/5	1.87	0.13	7%	
18, 13% Sb/Sn 270 nm	25	486	19	13	33/30	7/5	1.92	0.18	11%	
19, 12% Sb/Sn 440 nm	18	486	21	9.3	30/25	6/5	1.94	0.20	11%	
20, 9% Sb/Sn 445 nm	22	486	20	11	31/26	7/5	2.00	0.26	15%	

The selectivity is markedly increased, whereas the dominant transmitted wavelength, situated in the blue region, is slightly decreased.

EXAMPLES 21 to 25

In Examples 21 to 25, the process of Examples 1 to 5 is followed but
5 with “blue 2” glass as defined in Table 1, with a thickness of 4 mm:

[illegible]

Table 9:

Example	Blue 2 glass, 4 mm, LT/ET glass alone = 1.86									
	LTC	λ_p tr	P	ET	SF	LR	LT/ET	difference	% difference	
21, 5% Sb/Sn 280 nm	30	486	21	14	33/29	8/5	2.14	0.28	15%	
22, 9% Sb/Sn 285 nm	25	484	23	12	32/28	7.5/5	2.08	0.22	12%	
23, 13% Sb/Sn 270 nm	22	484	25	11	31/28	7/5	2.00	0.14	8%	
24, 12% Sb/Sn 440 nm	16	484	27	8	29/24	6/5	2.00	0.14	8%	
25, 9% Sb/Sn 445 nm	19	484	26	9.2	30/25	7/5	2.07	0.21	11%	

23

The selectivity is markedly increased, whereas the dominant transmitted wavelength remains identical or decreases.

EXAMPLES 26 and 27

For Example 26, the process of Example 1 is followed with "grey 1" glass as defined in Table 1. For Example 27, the glass is "grey 2" glass as defined in Table 1.

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Table 10:

	Grey 1 glass, 4 mm, LT/ET glass alone = 1.39 Grey 2 glass, 4 mm, LT/ET glass alone = 1.43									
	LTC	λ_0 tr	P	ET	SF	LR	LT/ET	difference	% difference	
Example 26										
Example 27										
26, 5% Sb/Sn 280 nm	28	495	6	15.9	30.3	5/7.1	1.76	0.37	27%	
27, 5% Sb/Sn 280 nm	37.9	496	5.2	21.3	34.6	5.7/4.4	1.78	0.35	24%	

The selectivity is very markedly increased with respect to the selectivity of the uncoated coloured glass.

EXAMPLES 28 to 43

These examples are presented in Tables 11 and 12. The coloured glasses used are mentioned in the second column of the table. The coloured glasses are those defined in Table 1. Furthermore, the blue glass "Azurlite" is used in Examples 42 and 43. This glass has the properties mentioned in Table 12. Its content of colouring agents is as follows:

0.41% Fe_2O_3 with 0.255% FeO ($\text{Fe}^{2+}/\text{total Fe}$ greater than 60%),
0.35-0.56% CeO_2
0.010% SO_3 .

The stackings of deposited layers are as follows:

- an underlayer of tin oxide with a thickness of 29 nm, then a layer, deposited by pyrolysis, based on tungsten oxide comprising 9.4% of Na with a thickness of 34 nm, covered with an SnO_2 layer with a thickness of 25 nm,
- an underlayer of tin oxide with a thickness of 16 nm, then a layer, deposited by pyrolysis, based on tungsten oxide comprising 9.4% of Na with a thickness of 78 nm, covered with an SnO_2 layer with a thickness of 40 nm,
- a "low emissivity" stacking comprising an SiO_x layer with a refractive index of 1.68 and a thickness of 70 nm coated with a low emissivity layer ($\epsilon = 0.15$) of tin oxide SnO_2 doped with fluorine (1 to 2% of fluorine) with a thickness of 320 nm. For the latter example, the values are measured with the layer in the 2 position, i.e. glass side, in order to benefit from the favourable effect of the low emissivity layer on the solar factor. In contrast, for all the other types of stackings, the values are measured with the layer in position 1 (coating side).

EXAMPLES 44 and 45

These examples are presented in Table 12. The coloured glasses used are the very dark green and dark green glasses defined in Table 1. The coating is formed of an absorbent stacking, with a thickness of the order of 75 nm, composed of a layer comprising essentially TiN and of a layer of SiC. The properties are measured with the layer in position 1 (coating side).

Table 11

Exam- ple		LTC	ET	SF	LT/ET	Diff.	% diff.	LT/SF	Diff.	% diff.	λ_D tr	P
	Support: blue 2 glass, 4 mm	39.9	21.4	40.1	1.86			0.99			486	24
28	GL/SnO ₂ (29)/WO ₃ Na9.4(34)/SnO ₂ (25)	37.4	18.7	29.7	2.00	0.14	8	1.26	0.27	28	485	21
29	GL/SnO ₂ (16)/WO ₃ Na9.4(78)/SnO ₂ (40)	21.1	10.5	18	2.0	0.15	8	1.17	0.19	19	484	24.7
30	GL/SiO _x 1.68(70)/SnO ₂ F(320) #2	37.2	19.7	31.9	1.89	0.03	2	1.17	0.18	18	485	19.6
	Support: green glass, 4 mm	67	38.6	53	1.74			1.27			504	3.5
31	GL/SnO ₂ (29)/WO ₃ /Na9.4(34)/ SnO ₂ (25)	62.8	33.1	41.5	1.90	0.16	9	1.51	0.25	20	497	5.2
32	GL/SnO ₂ (16)/WO ₃ Na 9.4(78)/SnO ₂ (40)	35.4	17.6	23.9	2.01	0.27	16	1.48	0.22	17	492	8.5

Table 11 cont.

Ex- am- ple		LTC	ET	SF	LT/ET	Diff.	% diff.	LT/SF	Diff.	% diff.	λ_D tr	P
	Support: very dark green glass, 4 mm	34.3	17.7	37.3	1.94			0.92			496	11.2
33	GL/SnO ₂ (29)/WO ₃ /Na9.4(34)/SnO ₂ (25)	32.2	15.2	26.8	2.12	0.18	9	1.20	0.29	32	494	12.8
34	GL/SnO ₂ (16)/WO ₃ Na9.4(78)/SnO ₂ (40)	18.1	8.1	16	2.23	0.29	15	1.13	0.22	25	492	15.8
	Support: dark green glass, 4 mm	47.5	25.2	42.9	1.88			1.11			498	7.7
35	GL/SnO ₂ (29)/WO ₃ /Na9.4(34)/SnO ₂ (25)	44.5	21.4	31.9	2.08	0.20	11	1.30	0.30	27	496	9.4
36	GL/SnO ₂ (16)/WO ₃ Na9.4(78)/SnO ₂ (40)	25.1	11.6	18.9	2.16	0.28	15	1.33	0.23	21	493	12.4
	Support: grey 2 glass, 4 mm	49.4	34.6	49.9	1.39			0.84			495	5
37	GL/SnO ₂ (29)/WO ₃ /Na9.4(34)/SnO ₂ (25)	46.3	26.7	36.3	1.73	0.30	21	1.28	0.29	29	492	6.9
38	GL/SnO ₂ (16)/WO ₃ Na9.4(78)/SnO ₂ (40)	26	13.8	20.8	1.88	0.45	32	1.25	0.26	27	489	10.3

Table 12

Exam ple		LTC	ET	SF	LT/ET	Diff.	% diff.	LT/SF	Diff.	% diff.	λ_D tr	P
	Support: grey 1 glass, 4 mm	36.5	26.3	43.7	1.39			0.84			494	5.8
39	GL/SnO ₂ (29)/WO ₃ Na9.4(34)/SnO ₂ (25)	34.2	20.4	31.1	1.68	0.29	21	1.10	0.27	33	492	7.7
40	GL/SnO ₂ (16)/WO ₃ Na9.4(78)/SnO ₂ (40)	19.2	10.6	18.1	1.81	0.42	30	1.06	0.23	28	489	11.1
41	GL/SiO _x 1.68(70)/SnO ₂ F(320) #2	34.1	24.1	35.6	1.41	0.03	2	0.96	0.13	16	493	5.3
	Support: blue glass, Azurlite (4 mm)	76.5	44	56.9	1.74			1.34			488	8.6
42	GL/SnO ₂ (29)/WO ₃ / Na9.4(34)/SnO ₂ (25)	71.8	39	46.4	1.84	0.10	6	1.55	0.20	15	487	10.4
43	GL/SnO ₂ (16)/WO ₃ Na 9.4(78)/SnO ₂ (40)	40.6	20.8	26.5	1.95	0.21	12	1.53	0.19	14	486	14
	Support: very dark green glass, 4 mm	34.3	17.7		1.94						496	11.2
44	GL/TiN/SiC	20.4	9.4		2.17	0.23	12				493	16.2
	Support: dark green glass, 4 mm	47.5	25.2		1.88						498	7.7
45	GL/TiN/SiC	28.2	13.2		2.14	0.26	14				494	12.9

CLAIMS

1. Glazing comprising:

- a substrate made of coloured soda-lime glass composed of main glass-forming constituents and of colouring agents, which exhibits a selectivity (LT/ET) of at least 1.1, measured with Illuminant C for a glass thickness of 4 mm,
- 5 - a pyrolytic coating deposited on the coloured glass substrate which provides the coated glazing with an increased selectivity with respect to the selectivity of the uncoated coloured glass.

2. Glazing according to Claim 1, characterized in that the selectivity of the uncoated coloured glass is at least 1.3.

10 3. Glazing according to Claim 1 or 2, characterized in that the selectivity of the uncoated coloured glass is less than or equal to 2.

4. Glazing according to any one of Claims 1 to 3, characterized in that the coloured glass is a glass for which the transmission between the wavelengths 1000 and 1200 nm, for a thickness of 4 mm, is lower by at least 5 points (expressed as %: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 500 and 600 nm.

5. Glazing according to any one of Claims 1 to 4, characterized in that the coloured glass is a soda-lime glass coloured dark grey composed of main glass-forming constituents and of colouring agents, in which glass the elements iron, selenium, cobalt and chromium are present as colouring agents in an amount corresponding to the following proportions (expressed as percentage by weight of the glass as if present in the form shown)

	Fe ₂ O ₃	0.75 to 1.8%
	Co	0.0040 to 0.0180%
25	Se	0.0003 to 0.0040%
	Cr ₂ O ₃	0.0010 to 0.0100%

and the proportions of the colouring agents are such that the glass exhibits a total energy transmission, measured for a thickness of 4 mm (ET₄), of between 15 and 40%, a selectivity (LTA/ET₄) of at least 1.2 and an excitation purity (P) not exceeding 10%.

30 6. Glazing according to any one of Claims 1 to 4, characterized in that the coloured glass is a green-coloured soda-lime glass which comprises the following percentages by weight of colouring agents, the total amount of iron being expressed in the form of Fe₂O₃:

30

Fe ₂ O ₃	0.7 to 1.3%
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FeO	0.18 to 0.27%
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Co	0 to 0.0040%
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V ₂ O ₅	0.0050 to 0.1%, and which exhibits,
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5 under Illuminant A and for a glass thickness of 4 mm, a light transmission (LTA4) of between 40 and 70% and a selectivity (LTA/ET4) of greater than or equal to 1.50.

7. Glazing according to any one of Claims 1 to 4, characterized in that the coloured glass is a grey-green soda-lime glass composed of main glass-forming constituents and of colouring agents which comprises less than 0.4% by weight of FeO and from 0.9 to 1.8% of Fe₂O₃, which has an excitation purity of more than 5% and which exhibits, under Illuminant A and for a glass thickness of 4 mm, a light transmission (LTA4) of greater than 30%, a selectivity (LTA/ET) of greater than 1.55 and an ultraviolet radiation transmission (UVT4) of less than 10%.

8. Glazing according to any one of Claims 1 to 4, characterized in that the coloured glass is a coloured soda-lime glass composed of main glass-forming constituents and of colouring agents which comprises from 0.40 to 0.52% by weight of FeO and which exhibits, under Illuminant A and for a glass thickness of 4 mm, a light transmission (LTA4) of less than 70%, a selectivity (LTA/ET4) of greater than 1.65 and an ultraviolet radiation transmission (UVT4) of less than 8%.

9. Glazing according to any one of the preceding claims, characterized in that the selectivity is increased by at least 3% with respect to the selectivity of the uncoated coloured glass.

10. Glazing according to any one of the preceding claims, characterized in that the selectivity is increased by at least 10% with respect to the selectivity of the uncoated coloured glass.

11. Glazing according to any one of the preceding claims, characterized in that the coating is a coating deposited by chemical vapour deposition.

12. Glazing according to any one of the preceding claims, characterized in that the coating is such that its transmission between the wavelengths 500 and 600 nm on clear glass with a thickness of 4 mm is higher by at least 3 points (expressed as percentage: ratio of the transmitted radiation to the incident radiation) with respect to the transmission between the wavelengths 1000 and 1200 nm.

13. Glazing according to one of the preceding claims, characterized in that the coating is chosen from the following:

- oxide coating deposited by pyrolysis comprising tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5,
- coating deposited by pyrolysis comprising a conductive or semiconductive layer with a thickness of 15 to 500 nm formed from a material comprising a metal oxide comprising a doping agent in a ratio of 5 to 100 mol per 100 mol of metal oxide, the metal oxide being selected from one or more of the following: tungsten oxide (WO_3), molybdenum trioxide (MoO_3), niobium pentoxide (Nb_2O_5), tantalum pentoxide (Ta_2O_5), vanadium pentoxide (V_2O_5) and vanadium dioxide (VO_2),
- coating deposited by pyrolysis which comprises an anti-reflective interferential stacking comprising, from the glass, a stacking of materials with alternatively high and low refractive indices,
- coating which comprises a layer with an emissivity of less than 0.3, in particular a layer deposited by pyrolysis based on fluorine-doped tin oxide,
- titanium nitride coating deposited by pyrolysis.

14. Glazing according to Claim 13, characterized in that the coating comprises an underlayer between the substrate and the coating deposited by pyrolysis.

15. Glazing according to Claim 13 or 14, characterized in that the coating is an oxide coating which is preferably deposited by vapour-phase pyrolysis comprises tin and antimony in a molar ratio Sb/Sn of between 0.04 and 0.16.

16. Glazing according to any one of Claims 13 to 15, characterized in that the coating is an oxide coating deposited by pyrolysis which comprises tin and antimony in a molar ratio Sb/Sn of between 0.01 and 0.5 and its thickness is between 250 and 500 nm.

17. Glazing according to any one of the preceding claims, characterized in that the coated substrate is bent and/or heat treated, in particular annealed or tempered.

18. Glazing according to any one of the preceding claims, characterized in that the light reflection factor (LR) is less than 13%.

19. Glazing according to any one of the preceding claims, characterized in that the emissivity is less than 0.2.

20. Glazing according to any one of the preceding claims, characterized in that the selectivity of the coated substrate is greater than 2.

21. Glazing according to any one of the preceding claims, characterized in that the dominant transmitted wavelength in the visible spectrum of

the coated substrate is less than the dominant transmitted wavelength of the uncoated substrate.

22. Glazing according to any one of the preceding claims, characterized in that the dominant transmitted wavelength in the visible spectrum of the coated substrate lies between 470 and 500 nm and the purity is less than 20%.

23. Glazing according to any preceding claim, in which the coating deposited on the coloured glass is such that if applied to 4 mm thick clear glass the so coated glass would have a light transmission factor measured with illuminant C of less than or equal to 70%.

24. Glazing according to any preceding claim, in which the coating deposited on the coloured glass is such that if applied to 6 mm thick clear glass the so coated glass would have a light transmission factor measured with illuminant C of less than or equal to 65%.

25. Glazing according to any preceding claim, in which the light transmission factor of the coated glazing is less than the light transmission factor of the uncoated glass substrate by a factor of greater than 10% calculated according to the formula

$$\frac{LT_{\text{uncoated substrate}} - LT_{\text{coated glazing}}}{LT_{\text{uncoated substrate}}} \times 100.$$

26. Glazing according to any preceding claim, in which the light transmission factor of the coated glazing is less than the light transmission factor of the uncoated glass substrate by a factor of greater than 15% calculated according to the formula

$$\frac{LT_{\text{uncoated substrate}} - LT_{\text{coated glazing}}}{LT_{\text{uncoated substrate}}} \times 100.$$

27. Use of the glazing according to any one of the preceding claims as glazing for a vehicle of the motor vehicle or train type.

Docket No.
P 63014 US 0

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

the specification of which

(check one)

☐ is attached hereto.

☒ was filed on 13 December 1999 as United States Application No. or PCT International Application Number PCT/EP99/09850 and was amended on 21 JUNE 2001

(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Not Claimed

<u>98124370.2</u>	<u>EP</u>	<u>22 DECEMBER 1998</u>	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	
<u> </u>	<u> </u>	<u> </u>	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	
<u> </u>	<u> </u>	<u> </u>	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

(Application Serial No.)

(Filing Date)

(Application Serial No.)

(Filing Date)

(Application Serial No.)

(Filing Date)

I hereby claim the benefit under 35 U. S. C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112, I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C. F. R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

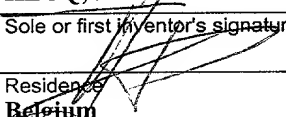
I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. *(list name and registration number)*


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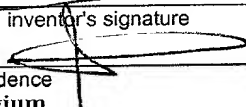
B-61

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US Patent

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IN RE APPLICATION OF: Andre HECQ et al.

ART UNIT:

SERIAL NO.: 09/868,848

EXAMINER:

INTERNATIONAL PCT/EP99/09850

APPLN. NO.:

FOR: COLOURED GLASS SUBSTRATE CARRYING A COATING

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